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Advances in Magnetism

Roadmap for Emerging Materials for Spintronic Device Applications

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The Technical Committee of the IEEE Magnetics Society has selected seven research topics to develop their roadmaps, where major developments should be listed alongside expected timelines: 1) hard disk drives; 2) magnetic random access memories; 3) domain-wall devices; 4) permanent magnets; 5) sensors and actuators; 6) magnetic materials; and 7) organic devices. Among them, magnetic materials for spintronic devices have been surveyed as the first exercise. In this roadmap exercise, we have targeted magnetic tunnel and spin-valve junctions as spintronic devices. These can be used, for example, as a cell for a magnetic random access memory and a spin-torque oscillator in their vertical form as well as a spin transistor and a spin Hall device in their lateral form. In these devices, the critical role of magnetic materials is to inject spin-polarized electrons efficiently into a nonmagnet. We have accordingly identified two key properties to be achieved by developing new magnetic materials for future spintronic devices: 1) half-metallicity at room temperature (RT) and 2) perpendicular anisotropy in nanoscale devices at RT. For the first property, five major magnetic materials are selected for their evaluation for future magnetic/spintronic device applications: 1) Heusler alloys; 2) ferrites; 3) rutiles; 4) perovskites; and 5) dilute magnetic semiconductors. These alloys have been reported or predicted to be half-metallic ferromagnets at RT. They possess a bandgap at the Fermi level E_F only for its minority spins, achieving 100% spin polarization at E_F . We have also evaluated $L1_0$ alloys and $D0_{22}$ -Mn alloys for the development of a perpendicularly anisotropic ferromagnet with large spin polarization. We have listed several key milestones for each material on their functionality improvements, property achievements, device implementations, and interdisciplinary applications within 35 years time scale. The individual analyses and the projections are discussed in the following sections.

Index Terms—Half-metallic ferromagnets, magnetic anisotropy, magnetic materials, spintronics.

I. HEUSLER ALLOYS

HEUSSLER alloys are ternary alloys originally discovered by Heusler [1]. He demonstrated the ferromagnetic behavior in an alloy consisting of nonmagnetic (NM) atoms, Cu_2MnSn . Since then, these alloys have been investigated due to their properties of shape memory and thermal conductance. In 1983, de Groot *et al.* [2] reported the half-metallic ferromagnetism in one of the Heusler alloys, half-Heusler NiMnSb alloy. A great deal of effort has accordingly been devoted to achieve the half-metallicity at room temperature (RT) using a Heusler alloy. In particular, Block *et al.* [3] measured a large tunneling magnetoresistance (TMR) in bulk full-Heusler $\text{Co}_2(\text{Cr, Fe})\text{Si}$ alloy, followed by a similar measurement in a thin-film form [4].

Among these Heusler alloys, Co-based full-Heusler alloys are the most promising candidates to achieve the RT half-metallicity due to their high Curie temperature

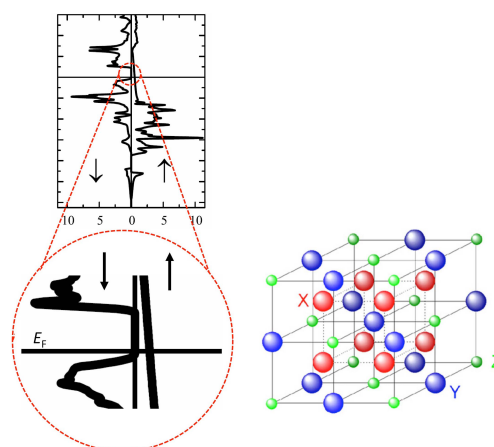


Fig. 1. Minority-spin bandgap [7] and $L2_1$ phase [6] of the full-Heusler alloys.

($T_C \gg \text{RT}$), good lattice matching with major substrates, large minority-spin bandgap (≥ 0.4 eV, see Fig. 1), and large magnetic moments in general [$\geq 4 \mu_B$ per formula unit (f.u.)] [5], [6]. The main obstacle to achieve the half-metallicity in the Heusler-alloy films is the vulnerability against the crystalline disorder, such as the atomic displacement, misfit dislocation, and symmetry break in the

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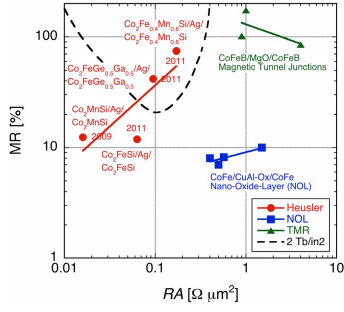


Fig. 2. Requirement for Tb/in² HDD read head and recent major results [9].

vicinity of the surface of the films. For the full-Heusler alloys forming X_2YZ , where the X and Y atoms are transition metals, while Z is either a semiconductor or an NM metal, the unit cell of the ideal crystalline structure ($L2_1$ phase, see Fig. 2) consists of four face-centered cubic sublattices. When the Y and Z atoms exchange their sites ($Y-Z$ disorder) and eventually occupy their sites at random, the alloy transforms into the $B2$ phase. In addition, the $X-Y$ and the $X-Z$ disorder finally leads to the formation of the $A2$ phase. By increasing the disorder, the magnetic properties depart further from the half-metallicity.

Toward the RT half-metallicity, two milestones have been identified as listed in the following:

- 1) (m1.1): demonstration of $>100\%$ giant magnetoresistance (GMR) ratio at RT;
- 2) (m1.2): demonstration of $>1000\%$ TMR ratio at RT.

Here, we have regarded these criteria using the MR as an indicator of the half-metallicity at RT.

Regarding (m1.1), in 2011, 74.8% GMR ratio was reported by Sato *et al.* [8] using a junction consisting of $\text{Co}_2\text{Fe}_{0.4}\text{Mn}_{0.6}\text{Si}/\text{Ag}/\text{Co}_2\text{Fe}_{0.4}\text{Mn}_{0.6}\text{Si}$. This is a significant improvement from 41.7% reported in [26]. Using such a GMR junction as a read head, the GMR ratio of $\sim 75\%$ with the resistance area product of $\sim 0.17 \Omega \cdot \mu\text{m}^2$ satisfies the requirement for 2 Tb/in² areal density in a hard disk drive (HDD). Fig. 2 shows the requirement and recent major efforts toward the Tb/in² areal density. It is clear that the Heusler-alloy GMR junctions are the only candidates satisfying the requirement to date. By reflecting on the development over the last five years, one can expect that the Heusler-alloy GMR junctions can achieve 100% GMR ratios within three years. This will satisfy (m1.1) and will lead to device applications as HDD read heads.

For (m1.2), Fig. 3 shows the development of the TMR ratios using amorphous and MgO barriers with both the conventional ferromagnets and the Heusler alloys as electrodes. As shown here, the largest TMR reported to date is 604% at RT using a magnetic tunnel junction (MTJ) of $\text{CoFeB}/\text{MgO}/\text{CoFeB}$ [10]. In 2005, an MTJ with an epitaxial $L2_1$ Co_2MnSi film has been reported to show very high TMR ratios of 70% at RT [11]. These are the largest TMR ratios obtained in an MTJ with a Heusler alloy film and an Al-O barrier. The TMR is purely induced by the intrinsic spin polarization of the Heusler electrodes, which is different from an MTJ with an oriented MgO barrier, where a TMR ratio of 386% has been achieved at RT (832% at 9 K) for $\text{Co}_2\text{FeAl}_{0.5}\text{Si}_{0.5}$ [12]. The TMR

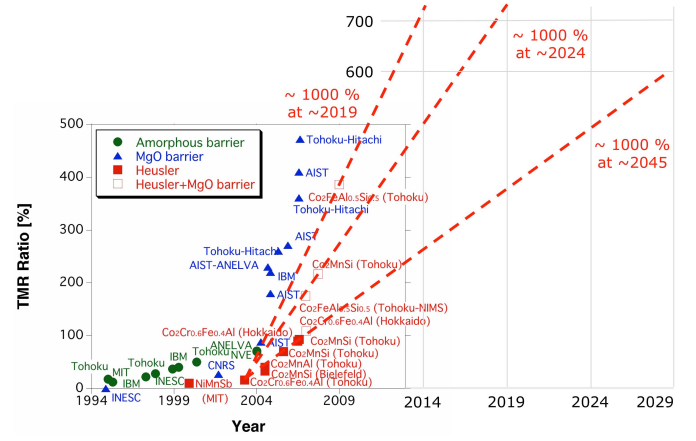


Fig. 3. Recent developments in the TMR ratios.

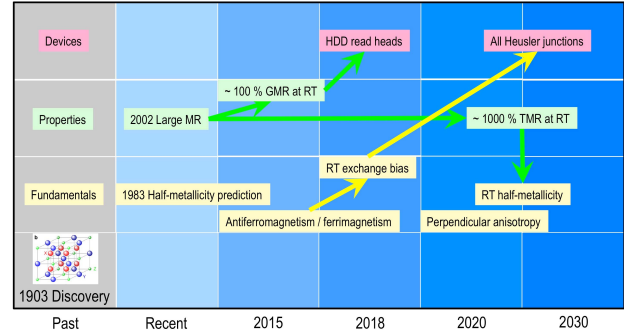


Fig. 4. Roadmap on the Heusler-alloy films.

ratio reported here is the highest ever in an MTJ with a Heusler alloy film but with the assistance of coherent tunneling through an oriented MgO barrier. By taking a moderate extrapolation, one can estimate that 1000% TMR ratios (m1.2) can be achieved within ten-year time period, i.e., the RT half-metallicity by 2024.

The other device application expected is to fabricate all Heusler junctions consisting of antiferromagnetic/ferromagnetic/NM/ferromagnetic Heusler-alloy layers. Such junctions can offer a template to avoid any crystalline disorder at the interfaces as the lattice matching and symmetry can precisely be controlled by atom substitution in these alloy layers. As a first step, Nayak *et al.* [13] reported an antiferromagnetic Heusler alloy of Mn_2PtGa for the first time but at low temperature (<160 K). One can anticipate that RT antiferromagnetism can be demonstrated within 5 years, leading to all Heusler-alloy junctions in 20 years.

By summarizing the above consideration, one can anticipate a roadmap on the Heusler-alloy films, as shown in Fig. 4. The Heusler-alloy films are expected to be used in GMR read heads and sensors within 3 years. These films are also to be combined with antiferromagnetic and/or NM Heusler-alloy films to form all Heusler junctions. Such junctions may be used in a magnetic random access memory subject to their perpendicular magnetic anisotropy, which is still in the infant stage in research.

II. OXIDES

Ferromagnetic oxide thin films have intensively been studied for more than last two decades due to their large

variety and tunability of physical properties, such as the ferro, ferri, antiferromagnetism, ferroelectricity, superconductivity, optical properties, and colossal MR (CMR) effect [14], [15]. In particular, some of ferromagnetic oxides are predicted as promising candidates of a half-metal and a spin filter, which directly lead to a large MR, as discussed in Section I. In addition, due to a high compatibility with other oxides and organic materials, the establishment of high-quality all-oxide heterostructure beyond CMOS device is highly expected. In this section, milestones and their associated roadmaps for three half-metallic oxide ferromagnets, (A) spinel ferrites, (B) rutiles, and (C) perovskites are discussed.

A. Spinel Ferrites

The most commonly studied oxides of Fe is Fe_3O_4 , which has an inverse spinel structure and a magnetic moment of $4.1 \mu_B/\text{f.u.}$ [16]. Among various spinel-type ferrites, Fe_3O_4 is a major conductive oxide at RT. The Curie temperature T_C is ~ 850 K, and the characteristic metal-insulator transition point (Verwey temperature) is 123 K. According to a band calculation, half-metallicity has been predicted [18], [19], and spin-resolved photoemission experiments show that Fe_3O_4 exhibits the spin polarization of up to -80% [20]. A very high spin polarization has also been suggested by the measurement of an MR ratio of over 500% through a nanocontact [21]. Epitaxial Fe_3O_4 films have been grown by various techniques, including molecular beam epitaxy under an oxygen atmosphere, magnetron sputtering, and laser ablation [20]. By replacing one of the Fe ions with a divalent metal ion, e.g., Mn, Co, Ni, and so on, a ferrite can be formed [20]. Siratori and Iida [22] have predicted half-metallicity in Mn, Co, and Ni ferrites, although the bulk materials are insulators except Fe_3O_4 . In particular, NiFe_2O_4 shows a bandgap in the majority band, indicating that this compound can become an insulator or semimetallic half-metal. The discrepancy of the bandgap structure between the *ab initio* calculation results and the experimental results suggests that the treatment of electron correlation is significant.

Some ferrites are expected as a good candidate of a spin filter because of their ferromagnetic insulator properties and high T_C . The spin-filtering device consists of a ferromagnetic insulator layer sandwiched between an NM metallic (NMM) layer and a ferromagnetic metallic (FMM) layer (or a superconductive layer). Due to the exchange splitting of the energy levels in the conduction band of the ferromagnetic insulator, the effective barrier height for the up-spin electron differs from that for the down-spin one, leading to a large difference in the tunneling probabilities between the two spin orientations. Therefore, ideally, an almost perfectly spin-polarized current is generated and this results in an infinite MR if a ferromagnetic insulator with a large exchange splitting is used. Here, the MR ratio is defined as $2P_{\text{SF}}P/(1 - P_{\text{SF}}P)$, where P_{SF} is the spin-filtering efficiency $[=(I_{\text{up}} - I_{\text{down}})/(I_{\text{up}} + I_{\text{down}})]$, $I_{\text{up(down)}} \propto \exp(-d \cdot \phi_{\text{up(down)}}^{1/2})$, where I is the tunneling current, d is the thickness of the spin filter, and ϕ is the effective barrier height and P is the spin polarization of the FMM layer. The RT spin-filtering effect has been demonstrated using

CoFe_2O_4 -based spin-filter devices [23], [24]. However, $|P_{\text{SF}}|$ at RT is $<5\%$.

Related to Section IV, the perpendicular magnetization behavior with a high uniaxial magnetic anisotropy of $K_u = 1.47 \times 10^6 \text{ J/m}^3$ in CoFe_2O_4 ferrite [25] has been reported. In addition to the ferromagnetic spinel ferrites, NM spinel, MgAl_2O_4 has also attracted much attention as a new spintronics material, because an ultrathin MgAl_2O_4 layer shows coherent tunneling properties (symmetry selective tunneling) and high MR ratios, such as an MgO tunnel barrier. Using an epitaxial $\text{CoFe/MgAl}_2\text{O}_4$ (with cation-site disordered)/CoFe structure, an MR ratio of $>300\%$ at RT was reported [26].

Toward the magnetic ferrites as a spintronic material, the following milestones have been recognized:

- 1) (m2.1.1): half-metallic behavior and high MR by improving the microstructure and the control of interface states;
- 2) (m2.1.2): high spin-filtering effects at RT by reducing structural and chemical defects;
- 3) (m2.1.3): tuning of perpendicular magnetic anisotropy;
- 4) (m2.1.4): development of new NM spinel-based materials to tune the transport properties and the coherent tunneling effect.

Regarding (m2.1.1) and (m2.1.2), ferrite films with a very high-quality crystalline structure, i.e., without any crystal imperfections, such as antiphase boundaries (APBs), atomic-site disorder, and dislocations, are necessary to obtain high saturation magnetization, high squareness of the hysteresis loops, and high T_C . The presence of APBs within a ferrite film, for instance, significantly degrades the saturation magnetization under a high magnetic field and the remanence. It also increases the resistivity of the film, since the APBs induce the electron-scattering center. Consequently, high-quality films are indispensable to the achievement of stable half-metallic characteristics and a spin-filtering effect at RT. In addition, the realization of a perfect and an abrupt ferrite/NM interface is required to preserve high effective spin polarization at the interface states. Therefore, the establishment of the growth method and procedures for the high-quality ferrite films, as well as a high-quality interface with the FMM layer and the NMM layer, are strongly desired. The development of an advanced growth process will lead to RT half-metallicity using ferrite family materials, such as Fe_3O_4 , $\gamma\text{-Fe}_2\text{O}_3$, CoFe_2O_4 , NiFe_2O_4 , MnFe_2O_4 , and ZnFe_2O_4 .

The milestone of (m2.1.3) is important to ensure the high thermal stability for nanoscale structures using CoFe_2O_4 -based ferrites for future spin-filtering devices and other spintronics use at RT. In particular, strong perpendicular magnetic anisotropy in a very thin region (below several nanometers) is desirable to control the tunneling resistance for device applications.

For (m2.1.4), providing the new NM tunnel barrier is now considered as an important issue to establish novel spintronic heterostructures, since only a limited tunnel barrier material (Al_2O_3 and MgO) is currently available to obtain high RT MR ratios. In particular, the ability to tune the physical properties is required to achieve higher performance, multifunctionality,

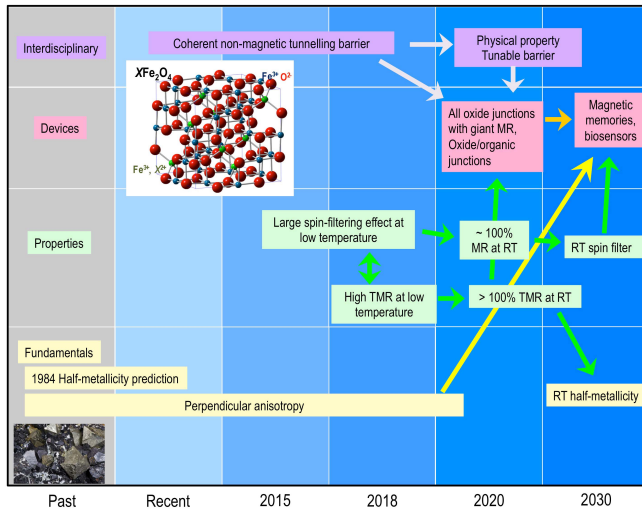


Fig. 5. Roadmap on the ferrite films.

and better compatibility to ferromagnetic electrodes. For instance, MR enhancement by crystalline barrier (coherent tunneling), a perfect lattice matching (lattice constant tuning), a low tunneling resistance (barrier height tuning), and applicability of high electric fields to a ferromagnetic layer facing the barrier (dielectric constant tuning) are presumably possible in spinel-based NM barrier with tailored compositions.

In summary, one can propose a roadmap on spinel ferrite films, as shown in Fig. 5. Using spinel ferrite-based MTJs consisting of ferrite/NM barrier/ferrite (or FMM) structure, $>100\%$ RT TMR (corresponding $|P|$ is ~ 0.7 according to the Julliere model) is expected within 10 years through the development of high-quality spinel ferrite thin films and the selection of a proper NM barrier. Further improvement of an MTJ structure and suppression of a rapid TMR reduction with increasing temperature will lead to a giant TMR over 1000% (corresponding $|P|$ is ~ 0.9) within 25 years.

To construct spin-filtering devices, one can use the techniques for the MTJ fabrication; a typical stacking structure is NMM/ferrite spin-filter/NM barrier/FMM, where the NM barrier is used to weaken the exchange coupling between the ferrite and the FMM layers. Recently, a higher P of -8% at RT (MR $\sim 6\%$) has been demonstrated using an epitaxial Pt/CoFe₂O₄/Al₂O₃/Co nanocontact junction [27]. Thus, the improvement of the junction structure as well as the ferrite film quality can enhance the MR ratio. More than 100% RT MR ratio due to the spin-filtering effect is expected within 10 years by reducing structural and chemical defects in spin-filter junctions.

Using new NM barriers, one can highly expect a giant TMR ratio exceeding 500% at RT within 5 years. Furthermore, the tuning of physical properties will be achieved by searching for new candidate barrier materials within 10 years.

B. Rutiles

Using Andreev reflection, CrO₂ has been proven to show a half-metallic nature at low temperature, as suggested by the *ab initio* calculations [16], [17]. The high spin polarization of 90% has been confirmed at low

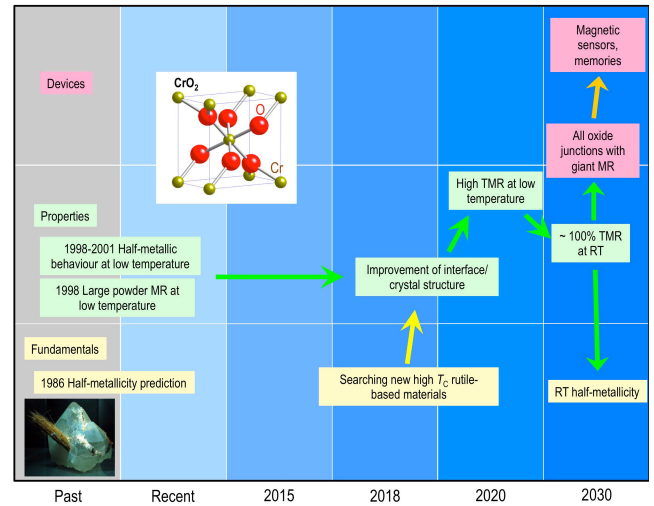


Fig. 6. Roadmap on the rutile films.

temperature using the point-contact Andreev reflection method [18], [19], and high powder MR has been reported [20]. However, RT half-metallicity has not been demonstrated yet. CrO₂ has a tetragonal unit cell with a magnetic moment of $2.03 \mu_B/\text{f.u.}$ at 0 K [21]. The ferromagnetism of CrO₂ appears <391 K [22]. Above this temperature another phase of Cr₂O₃ is known to show antiferromagnetism, which is the major cause of the reduction of the half-metallicity. Highly ordered CrO₂ films are predominantly grown by chemical vapor deposition [23]. However, obtaining the CrO₂ single phase as a thin film is not easy, and thus MR properties steeply decrease below RT.

In order to utilize the rutiles in a spintronic device, the following milestones have been identified:

- 1) (m2.2.1): development of a high-quality CrO₂ thin film with a single rutile phase and achievement of a clean interface structure with tunnel junctions;
- 2) (m2.2.2): search for new rutile-based materials with higher T_C and robust half-metallicity by tailoring their composition.

Regarding (m2.2.1), the undesirable reduction in MR ratio below T_C could be suppressed by the improvement of the crystal structure and the interface state. The optimization of an epitaxial growth process for a single rutile phase and the use of a suitable NM barrier, which does not invade the interface of CrO₂, will be effective. In addition, the elimination of the NM Cr₂O₃ phase, which generally forms on the surface of the CrO₂ film, using sophisticated deposition and treatment processes will enhance the magnetic and half-metallic properties.

For (m2.2.2), to obtain a more stable half-metallic phase with high T_C , doping of other elements to CrO₂ or searching ternary or quaternary rutile-based ferromagnetic materials would be necessary. Such a new composition and a new material will lead to stable half-metallic properties and higher MR at RT.

In summary, one can anticipate a roadmap on the half-metallic rutile films, as shown in Fig. 6. Obtaining epitaxial thin films with a single CrO₂ phase will lead to the observation

of RT TMR ratios within 10 years. To demonstrate high TMR ratios ($>100\%$) at RT is still challenging. Searching new rutile-type ferromagnetic oxides and a sophisticated MTJ structure might yield a technological breakthrough toward a higher TMR ratio in the future.

C. Perovskites

Perovskites, such as $(\text{La}, \text{Sr})\text{MnO}_3$, exhibit both strong ferromagnetism and metallic conductivity with a partial substitution of La^{+3} ions with 2^+ ions, such as Ca, Ba, Sr, Pb, and Cd [28], [29]. Since only one spin band exists at E_F in these films, 100% spin polarization can be achieved. Using these materials instead of a conventional ferromagnet, a very high MR of $\sim 150\%$ at RT has been observed [30]. This is known as CMR. Using Mn-perovskite thin films and SrTiO_3 oxide tunnel barrier, a TMR ratio of up to 1850% has been reported but only below T_C [31]. CMR can be induced either by breaking the insulating symmetry of Mn^{3+} and Mn^{4+} alternating chains or by suppressing spin fluctuation near T_C . Even so, it is unlikely to achieve the RT half-metallicity in the conceivable future.

Much effort has been spent to search for new high T_C perovskites for an RT half-metallicity. The family of double perovskites with a chemical composition of $\text{A}_2\text{BB}'\text{O}_6$ (A is an alkaline earth or rare-earth ion, B and B' are transition metal ions) has been focused for more than 15 years, since some of the double perovskites exhibit high T_C above RT and half-metallic band structures [32]. $\text{Sr}_2\text{FeMoO}_6$ (SFMO) has high T_C of 420 K and has been predicted to be a half-metal [33], indicating the double perovskites are a promising oxide family for high MR at RT. At low temperature, high $P \sim -80\%$ in an SFMO film has been demonstrated using a $\text{Co}/\text{SrTiO}_3/\text{SFMO}$ MTJ. Much higher T_C of 635 K is reported in $\text{Sr}_2\text{CrReO}_6$ [34].

Recently, 2-D electron gas (2-DEG) at the interface of an NM perovskite heterostructure consisting of $\text{LaAlO}_3/\text{SrTiO}_3$ has intensively been investigated due to a high mobility in the 2-DEG. Highly efficient spin transport in the 2-DEG could be usable to establish the new type spin transistors in the future.

The following milestones have been established toward the perovskites as a spintronic material:

- 1) (m2.3.1): search for new perovskite-based materials with $T_C > \text{RT}$;
- 2) (m2.3.2): development of a high MR at RT.

Regarding (m2.3.1), the double perovskites with A_2FeMoO_6 or A_2FeReO_6 series are promising due to their high T_C . However, a high MR using an MTJ structure has not been achieved, since there are some considerable obstacles against (m2.3.2): 1) site disorder of magnetic ions deteriorates the magnetic properties and the spin polarization and 2) their high reactivity to water, which restricts the use of common microfabrication techniques.

In order to overcome these obstacles, the improvement of film quality and the preparation of a clean interface are necessary to achieve the high MR ratios at RT. In particular, the specific microfabrication method should newly be developed

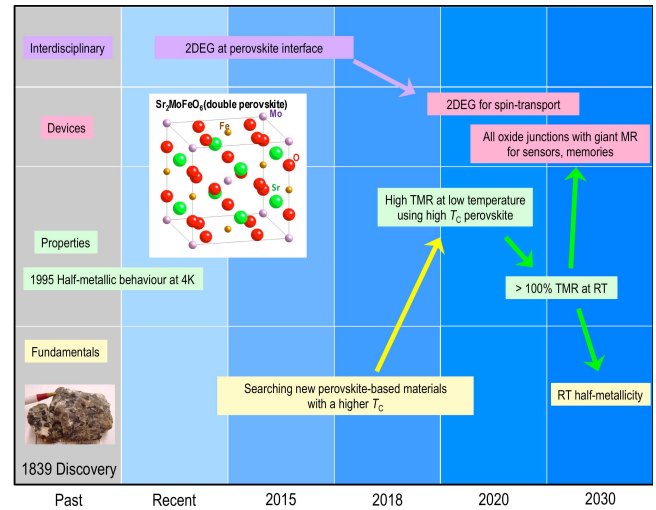


Fig. 7. Roadmap on the perovskite films.

to reduce the damage during the processes. In addition, a new barrier material that matches with the perovskites will be needed to compose a high-quality perovskite-based MTJ.

In summary, one can expect a roadmap on the perovskite films, as shown in Fig. 7. RT TMR ratios will be obtained using the MTJs with a high T_C perovskite layer within 5 years. Less than 100% TMR at RT will be expected in the future after the demonstration of high TMR ratios at low temperatures.

III. DILUTE MAGNETIC SEMICONDUCTORS

Unlike metals, semiconductors have a relatively low carrier density that can drastically be changed by doping, electrical gates, or photoexcitations, to control their transport and optical properties. This versatility makes them the materials of choice for information processing and charge-based electronics. In magnetically doped semiconductors, such as $(\text{Cd}, \text{Mn})\text{Te}$, $(\text{In}, \text{Mn})\text{As}$, or $(\text{Ga}, \text{Mn})\text{As}$, these changes of carrier density also enable novel opportunities to control the magnetic properties and lead to applications that are not available or ineffective with ferromagnetic metals [35]. For example, a carrier-mediated magnetism in semiconductors offers a tunable control of the exchange interaction between the carriers and the magnetic impurities. The onset of ferromagnetism and the corresponding change in the T_C can be achieved by increasing the carrier density using an applied electric field, photoexcitations, or even heating. Two milestones for the research on novel magnetic semiconductors are identified:

- 1) (m3.1): search for tunable ferromagnetism in semiconductors with $T_C > \text{RT}$.
- 2) (m3.2): demonstrating RT devices that are not limited to magnetoresistive effects.

Considering (m3.1), despite numerous reports for $T_C > 300$ K in many semiconductors, a reliable RT ferromagnetic semiconductor remains elusive [36], [37]. However, even the existing low- T_C magnetic semiconductors have provided the demonstrations of novel magnetic effects and ideas that have also subsequently been transferred to ferromagnetic metals, for example, electric-field modulation of coercivity and magnetocrystalline anisotropy at RT [37].

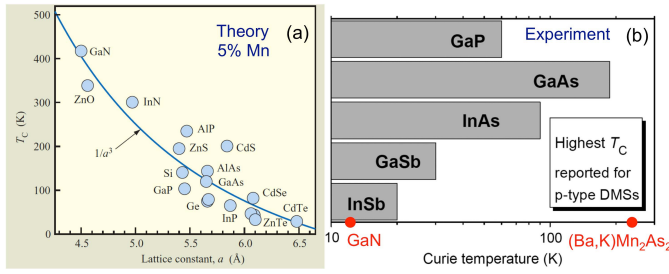


Fig. 8. (a) Theoretical predictions for T_C in DMS [41], adapted from [45]. (b) Reliable highest experimental T_C reported for Mn-doped DMS, adapted from [36].

An early work on ferromagnetic semiconductors dates back to CrB_3 in 1960 [38]. Typically studied were concentrated magnetic semiconductors, having a large fraction of magnetic elements that form a periodic array in the crystal structure. Important examples are Eu-based materials, in which the solid-state spin-filtering effect was demonstrated for the first time [39]. However, the complicated growth and the modest T_C (up to ~ 150 K) limited these materials to fundamental research. Starting with the mid-1970s, the dilute magnetic semiconductors (DMS), alloys of NM semiconductor and magnetic elements (typically, Mn) [40], became intensely explored first in II–IV, and later in III–V NM hosts. In II–VIs, Mn^{2+} is isovalent with group II providing only spin doping, but not carriers and thus making robust ferromagnetism elusive. In III–Vs, Mn yields both spin and carrier doping, but low-Mn solubility limit complicates their growth and can lead to an extrinsic magnetic response due to nanoscale clustering of metallic inclusions. This complex dual role of Mn doping in III–Vs possess both: 1) challenges to establish the universal behavior among different NM III–V hosts. $(\text{Ga}, \text{Mn})\text{N}$ predicted to have $T_C > 300$ K [41], but shown to only have $T_C \sim 10$ K [42] and 2) makes the *ab initio* studies less reliable, requiring careful considerations of secondary phases and magnetic nanoclustering—a source of many reports for an apparent high- T_C in the DMS.

An important breakthrough came with the growth of III–V DMS: $(\text{In}, \text{Mn})\text{As}$ in 1989 and $(\text{Ga}, \text{Mn})\text{As}$ in 1996 [43], [44], responsible for demonstrating tunable T_C , coercivity, magnetocrystalline anisotropy, as well as the discovery of tunneling anisotropic MR [37]. However, even if the low-Mn solubility is overcome (maximum $\sim 10\%$), the upper T_C limit is given MnAs with $T_C \sim 330$ K. This suggests that $(\text{Ga}, \text{Mn})\text{As}$, with the current record $T_C \sim 190$ K [41], is not a viable candidate for RT ferromagnetism in DMS. Influential mean-field calculations [39] for DMS with 5% Mn in Fig. 8(a) show a strong correlation with an inverse unit cell volume [45]. However, the *ab initio* studies reveal a more complex, material-dependent situation [46].

Instead of III–V compounds, more promising is recently discovered II–II–V DMS [47]. They are isostructural to both 122 class of high-temperature Fe-based superconductors and antiferromagnetic BaMn_2As_2 , offering intriguing possibilities to study their multilayers with different types of ordering. In $(\text{Ba}, \text{K})(\text{Zn}, \text{Mn})_2\text{As}_2$ with an independent carrier (K replacing Ba) and spin doping (Mn replacing Zn),

some of the previous limitations are overcome: the absence of carriers in II–VIs and the low-Mn solubility in III–Vs. With 30% K and 15% Mn doping, their $T_C \sim 230$ K [48] exceeds the value in $(\text{Ga}, \text{Mn})\text{As}$. Selected highest reliable experimental T_C reported for the Mn-doped DMS is shown in Fig. 8(b). Circles are given for GaN, which has about 30 times smaller T_C than predicted in Fig. 8(a), and $(\text{Ba}, \text{K})\text{Zn}_2\text{As}_2$, a current record for DMS. The *ab initio* studies predict a further increase in T_C [49]. We expect that the tunable RT carrier-mediated ferromagnetism will be realized in II–II–V DMS within 5 years.

Regarding (m3.2), while DMS is often viewed as the materials for multifunctional devices to seamlessly integrate nonvolatile memory and logic [35], other device opportunities could be more viable. DMS-based optical isolators [50], [51] were already commercialized by Tokin Corporation [52]. Such devices, relying on large magneto-optical effects (Faraday and Kerr) that are proportional to the giant Zeeman splitting in DMS, are used to prevent feedback into laser cavities and provide a one-way transmission of light. Even without demonstrating $T_C > \text{RT}$, enhancing RT Zeeman splitting is important for DMS (exceeding a large g -factor ~ 50 for InSb).

Spin lasers [53], [54] are another example of devices not limited to MR effects. They can outperform [55], [56] conventional lasers with injected spin-unpolarized carriers. For spin lasers, electrical spin injection is desirable, currently limited up to ~ 230 K [57]. $T_C > \text{RT}$ in DMS would be beneficial to such spin lasers, both as an efficient spin injector and possibly a tunable active region that could alter the laser operation through the tunable exchange interaction. To remove the need for an applied B -field, the perpendicular anisotropy of the spin injector is suitable. We expect RT electrical spin injection in spin lasers by 2020. It is important to critically assess if extrinsic $T_C > \text{RT}$ in DMS, from magnetic metallic nanoinclusions and secondary phases [having $\text{GaAs} + \text{MnAs}$, rather than $(\text{Ga}, \text{Mn})\text{As}$, a true DMS] is a viable path for RT spintronic devices. RT magnetoamplification was demonstrated in $(\text{In}, \text{Mn})\text{As}$ -based magnetic bipolar transistor, operating above $T_C < 100$ K of a single-phase $(\text{In}, \text{Mn})\text{As}$ [58]. Another test for useful extrinsic (multiphase) DMS is a robust RT electrical spin injection. A road map for DMS is shown in Fig. 9.

IV. PERPENDICULARLY ANISOTROPIC FERROMAGNETS

A perpendicularly magnetized system is currently an important building block in spintronic devices, since it enables us to shrink the size of memory bits and to reduce the electric current density required for spin-transfer switching. There are several ways to obtain the perpendicular magnetic anisotropy in a thin film. To use an ordered alloy showing high magnetocrystalline anisotropy is one possible way. If its easy magnetization axis is aligned along the normal direction to the film plane, and the magnetocrystalline anisotropy field overcomes the demagnetization field, the film shows the perpendicular magnetization. Another way is to use the

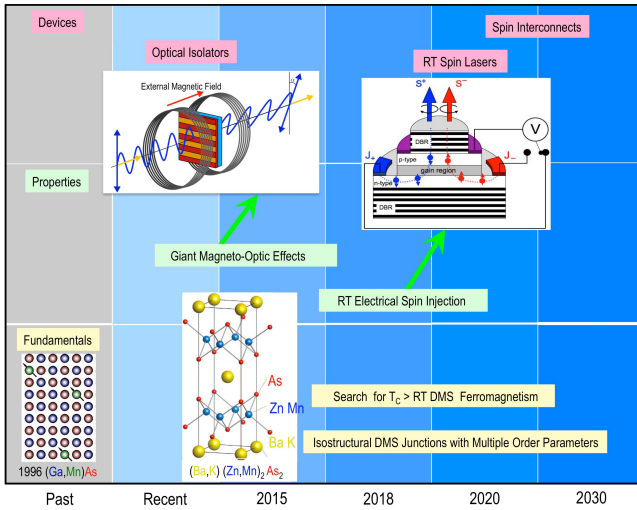


Fig. 9. Roadmap on DMSs.

interface magnetic anisotropy between a ferromagnetic layer and an NM layer. In addition, multilayered structures are useful to obtain perpendicular magnetization.

Toward the perpendicularly anisotropic ferromagnet as a spintronic material, the following milestones have been established:

- 1) (m4.1): high thermal stability of perpendicular magnetization;
- 2) (m4.2): structural stability against the thermal process;
- 3) (m4.3): demonstration of the high spin polarization;
- 4) (m4.4): reduction of the magnetic damping constant.

(m4.1) means the stability of magnetization at a nanometer scale overcoming the magnetization fluctuation due to the thermal energy. Considering several thermal treatments in device fabrication processes, (m4.2) should be satisfied. (m4.3) is a key determining the performance of MTJ and GMR devices. In terms of spin-transfer torque (STT) magnetization switching, as indicated in (m4.4), the magnetic damping should be small to reduce the electric current density for switching.

An $L1_0$ -ordered structure exists in the thermodynamically stable phase and is composed of the alternative stacking of two kinds of atomic planes along the c -axis. Thus, $L1_0$ -ordered alloys, such as FePt, FePd, CoPt, MnAl, and MnGa, exhibit uniaxial magnetic anisotropy along the c -axis direction. When one aligns the c -axis of $L1_0$ -ordered structure in the normal direction to the film plane, a perpendicular magnetic anisotropy is obtained. Since the $L1_0$ -ordered structure is thermally stable, $L1_0$ -ordered alloys have an advantage from the viewpoint of (m4.2). Among the $L1_0$ -ordered alloys, $L1_0$ -FePt shows the largest uniaxial anisotropy (K_u) of 7×10^6 J/m³ [59], which leads to the excellent thermal stability of magnetization at a reduced dimension, e.g., 4 nm diameter in $L1_0$ -FePt nanoparticles. This property satisfies (m4.1). Because of its perpendicular magnetization for FePt (001) films, $L1_0$ -FePt has been regarded as an ideal material for perpendicular recording media in an HDD. In addition, the spin polarization of FePt is theoretically predicted to be approximately 70% [24], which is a good

characteristic for a spintronic material. $L1_0$ -ordered FePt films have already been implemented in both the MTJ [60] and the GMR [24] junctions. In the case of GMR nanopillars consisting of two FePt layers separated by NM Au, the STT phenomena have systematically been examined by tuning the crystalline order of the FePt layer [24]. However, the observed TMR and the GMR ratios are still low for $L1_0$ -FePt.

Another important issue is that the major $L1_0$ -ordered alloys contain the heavy transition metals, such as Pt. The Pt atom shows strong spin-orbit coupling, which leads to the significant enhancement of magnetization damping. This feature is an opposite trend to (m4.4). $L1_0$ -FePd exhibits a large K_u and rather smaller damping constant compared with that of $L1_0$ -FePt, probably because Pd is lighter element than Pt [61]. However, the usage of such noble metals as Pt and Pd is not suitable from the viewpoint of element strategic trend. Considering these recent demands, a new kind of $L1_0$ alloy is eagerly desired, which possesses a large K_u and a small damping constant. One of the candidates is $L1_0$ -FeNi. Since a paper reported that an $L1_0$ -FeNi bulk alloy exhibited high uniaxial magnetic anisotropy of $K_u = 1.3 \times 10^6$ J/m³ [62], $L1_0$ -FeNi is a future material having a possibility to substitute high K_u materials containing the noble metals and rare earths. Kojima *et al.* [63] reported the preparation of $L1_0$ -FeNi thin films with a relatively high K_u of 0.7×10^6 J/m³, and also the small damping constant has been reported in $L1_0$ -FeNi [64].

Another candidate material showing perpendicular magnetization is an Mn-based alloy system, such as $L1_0$ -MnAl. Recently, epitaxial Mn-Ga films, including $L1_0$ - and $D0_{22}$ -ordered phases, have also been found to exhibit strong perpendicular magnetic anisotropy ($K_u = 1.2$ – 1.5×10^6 J/m³) with small saturation magnetization ($M_s = 250$ – 500 emu/cm³) and small magnetic damping ($\alpha = 0.0075$ – 0.015) at RT [65], [66]. Moreover, it has been found that $D0_{22}$ -Mn₃Ge epitaxial films exhibited K_u of 0.91×10^6 J/m³ [67] and 1.18×10^6 J/m³ [68]. These Mn-based alloy systems can also be used as a perpendicular magnetized layer for STT application, because the *ab initio* calculations predicted the high spin polarization of 88% for Mn₃Ga [69] and a half-metallic band dispersion for Mn₃Ge that leads a high TMR, such as Fe/MgO-MTJs [70], [71]. However, the observed TMR ratios are also still low for $L1_0$ -Mn-Ga and $D0_{22}$ -Mn-Ga [72]. Experimental realization of the high spin polarization is essential for all the ordered alloys to achieve (m4.3).

Multilayered structures, such as Co/Pt, Co/Pd, Co/Ni, and so on, also show the perpendicular magnetization. The main origins for perpendicular magnetic anisotropy in the multilayered structures are as follows: 1) breaking the crystal symmetry at the interface, which leads to the interface magnetocrystalline anisotropy; 2) the effect of magnetostriction due to the interface between different atomic planes; and 3) interface alloying. Although the multilayered films show high magnetic anisotropy, we need to consider the stability of the layered structure against a thermal process. In some cases, the high temperature annealing degrades the layered structure

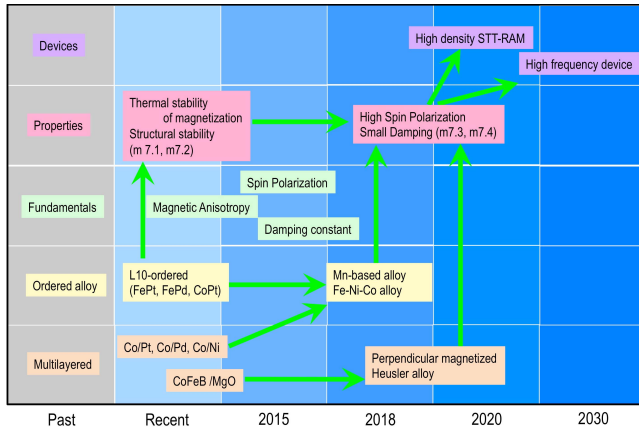


Fig. 10. Roadmap on the perpendicularly anisotropic films.

and its magnetic properties, which should be improved for (m4.2). Mangin *et al.* [73] and Meng and Wang [74] also demonstrated the STT switching in CPP-GMR nanopillars with perpendicularly magnetized Co/Ni and Co/Pt multilayers, respectively. As in the case of the ordered alloys, however, increasing MR effect and lowering magnetization damping are inevitable issues for the multilayered structures to achieve (m4.3) and (m4.4). To explore the adequate materials combination is one of the ways for the multilayered structure to solve the current problems.

One of the new types of multilayering films is an artificial superlattice grown using nearly monoatomic layer alternation of Co and Pt or Pd. Such ultrathin superlattice films had an annealing stability higher than that of the conventional multilayering films [75].

It has also been reported that the CoFeB/MgO junction shows perpendicular magnetic anisotropy [76]. The perpendicular magnetization components of the CoFeB are induced at the MgO interface, which originates from the interface magnetic anisotropy. The perpendicularly magnetized CoFeB/MgO layers have a significant advantage, because MgO-based tunnel junctions show a high TMR ratio. Actually, it has also been demonstrated that a CoFeB/MgO/CoFeB stack with perpendicular magnetization shows the TMR ratio over 120% and the low STT switching current of 49 μ A at a 40 nm-diameter junction. This is a promising candidate as a building block for the MRAM cell. However, because the interfacial magnetic anisotropy constant is not large enough, and a thin ferromagnetic layer is required to exploit the interface effect, the small volume of the magnetic layer may give rise to the thermal instability of magnetization in a deeper subnanometer region. (m4.1) is an important step for the perpendicular anisotropic ferromagnets using the interface magnetic anisotropy. In addition, perpendicularly magnetized Heusler alloy layers, where interface magnetic anisotropy is used, are attracting attention as an alternative perpendicularly magnetized system, which may lead to the high spin polarization (m4.3) and a low damping constant (m4.4). Recently, the perpendicular magnetization and the TMR ratio of 132% at RT have been demonstrated using an ultrathin Co₂FeAl Heusler alloy/MgO/CoFeB MTJ [77]. These are summarized in Fig. 10.

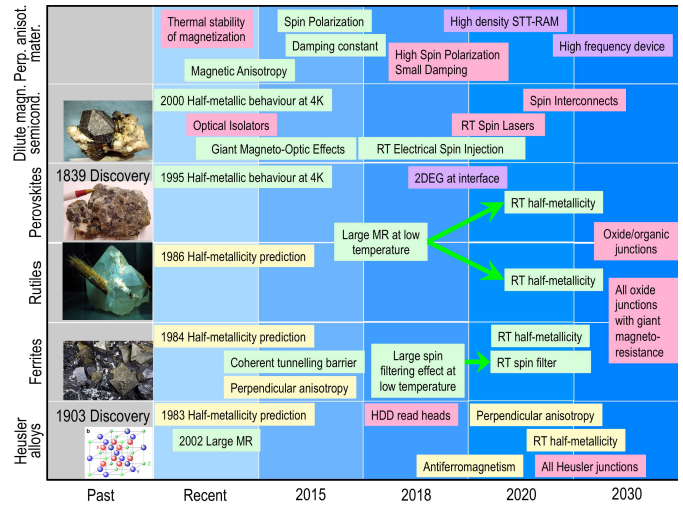


Fig. 11. Roadmap for magnetic materials.

V. OVERVIEW

In this roadmap, we have identified two key properties to develop new (and/or improved) spintronic devices. The first one is the half-metallicity at RT, which can be achieved by clearing milestones to realize large MR and resulting large spin polarization. The second one is the perpendicular anisotropy in nanoscale devices at RT. This is based on milestones, including large perpendicular magnetic anisotropy and small damping constant. Such development is expected to be achieved not only by the development of these alloys but also by the fundamental understanding on these properties using a well-studied test system, i.e., zincblendes. As summarized in Fig. 11, we anticipate these materials investigated here to realize all Heusler and all oxides junctions. These can be implemented in the next-generation MRAM and high-frequency devices within 35 years.

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